Low temperature Kondo reduction of quadrupolar and magnetic moments in the YbCu_5 − xAl_x series

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The variational solution of the Kondo problem applied to the $YbCu_{5-x}Al_x$ series

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Abstract

We present measurements in the $YbCu_{5-x}Al_x$ series, down to the 50 mK range, using ^{170}Yb Mössbauer absorption spectroscopy and magnetisation measurements. In this series, the hybridisation between the Yb $4f$ electrons and the conduction electrons is known to decrease as the Al content x increases. We apply the variational solution of the impurity Kondo problem to the interpretation of our data. We show that the Kondo temperature can be derived from the measured $4f$ quadrupole moment and, for the magnetically ordered compounds ($x \geq 1.6$), we obtain the exchange energy as a function of the Al content. Our findings are in general agreement with Doniach's model describing the onset of magnetic ordering according to the relative values of the Kondo and exchange energy scales.

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I. INTRODUCTION

In previous works ([?] and references therein), it was shown that a progressive substitution of Al for Cu in $YbCu_5$ leads to a decrease of the hybridisation between the Yb $4f$ electrons and the metal electrons: the Yb ion has an intermediate valence in $YbCu_5$ whereas it is nearly trivalent in $YbCu_3Al_2$. Near a critical Al content $x_c \simeq 1.5$, the state with magnetic ordering of the Yb ions becomes the more stable. Concomitantly, the specific heat data for compounds around this critical concentration reveal the occurence of a Non-Fermi liquid behaviour ($1.3 \leq x \leq 1.6$). The valence v of the Yb ion, for $x \geq 1.3$, has been determined by L_{III}-edge x-ray absorption to be very close to 3, i.e. the number of electrons in the $4f$ shell $n_f = v - 2$ is close to 0.98. The $YbCu_{5-x}Al_x$ series for $x \geq 1.3$ appears therefore as a good candidate for testing existing theories describing the ground state of a Kondo lattice, where a weak $4f$-band electron hybridisation is present [?].

In the present work, we will be interested in the T=0 properties of the Yb^{3+} ion in the $YbCu_{5-x}Al_x$ series for $x \geq 1.3$, obtained by ^{170}Yb Mössbauer absorption spectroscopy and magnetisation measurements down to temperatures in the 50 mK range. The theoretical background is that of the T=0 variational solution of the Kondo impurity problem, as developed by Gunnarsson and Schönhammer [?] and applied to the crystal electric field (CEF) split levels of the Yb^{3+} ion in [?].

II. THE VARIATIONAL APPROACH TO THE KONDO IMPURITY PROBLEM

The variational solution of the Anderson hamiltonian describing a paramagnetic ion hybridised with conduction electrons [?] is derived by minimizing the energy of the ground N-electron state represented by the trial function:

$$|\psi\rangle = A[|\Omega\rangle + \frac{1}{N_f} \sum_{m,k<k_F} a_{km} f_m^\dagger c_{km} |\Omega\rangle]. \quad (1)$$

In this expression, A and the a_{km} are coefficients, $|\Omega\rangle$ is the vacuum state made of the full Fermi sea with an "empty" $4f$ orbital (no $4f$ electron in the case of Ce^{3+}, no $4f$ hole in the

case of Yb^{3+}), with degeneracy N_f, $f_m^\dagger$ is the creation operator of a $4f$ state with magnetic quantum number m, and c_{km} is the annihilation operator of a conduction electron with quantum numbers k and m. The $4f$ states $|m\rangle$ are the eigenvectors, with energy Δ_m, of the crystal electric field interaction, to which is added an exchange or Zeeman interaction in the case of magnetic ordering or of application of an external magnetic field respectively. The mean value of an operator O acting on the $4f$ variables, in the ground state $|\psi\rangle$, i.e. at T=0, can be written as [**?**]:

$$\langle O\rangle = \sum_m n_{fm}\langle m|O|m\rangle, \tag{2}$$

where the n_{fm} are $4f$ "occupation numbers" given by:

$$n_{fm} = \frac{\Gamma(1-n_f)}{\pi}\frac{1}{T_K+\Delta_m}. \tag{3}$$

In this expression, $n_f = \sum_m n_{fm}$ is the electron count in the $4f$ orbital, which is close to 1 in the Kondo limit of weak hybridisation; T_K is the single ion Kondo temperature of the non-degenerate $4f$ electrons; and Γ is the bare hybridisation width linked to n_f and T_K by the relationship, obtained by summing Eqn(**??**) over m:

$$\sum_m \frac{1}{T_K+\Delta_m} = \frac{\pi n_f}{\Gamma(1-n_f)}. \tag{4}$$

A consequence of the presence of hybridisation, i.e. of a N-electron ground state containing a small admixture of the vacuum state $|\Omega\rangle$ with an empty $4f$ orbital, is that the T=0 mean value of the observable O is modified with respect to its "hybridisation free" T=0 value. For the magnetic moment $m = -g_J J_z \mu_B$ (for Yb^{3+}: $g_J = 8/7$, $J = 7/2$), Eqn.(**??**) always leads to a hybridised T=0 moment smaller than the pure CEF moment, for a Kramers ion like Yb^{3+} or Ce^{3+}: this is the well-known magnetic moment reduction due to the Kondo screening. For the case of the $4f$ quadrupole moment $Q_{zz} = 3J_z^2 - J(J+1)$, the T=0 value of $\langle Q_{zz}\rangle$ in the presence of hybridisation depends on the crystal field level scheme of the Yb^{3+} ion. In the case where the $4f$ quadrupole moment of the ground CEF state is large and positive, it can be shown that Eqn.(**??**) leads to a reduction of the T=0 $\langle Q_{zz}\rangle$ value

with respect to the CEF value. These two quantities can be measured by ^{170}Yb Mössbauer spectroscopy: the magnetic moment m is obtained in the magnetically ordered phase from the hyperfine field at the ^{170}Yb nucleus; and the principal component Q_{zz} of the quadrupole moment tensor is obtained in the paramagnetic phase from the electric field gradient at the nucleus.

III. THE MÖSSBAUER DATA AND THE VARIATIONAL CALCULATION IN THE PARAMAGNETIC PHASE OF THE YBCU$_{5-X}$AL$_X$ SERIES

The site symmetry of the Yb atom in the $YbCu_{5-x}Al_x$ series is very close to hexagonal (see for instance Ref. [**?**]). At 4.2 K and above, i.e. in the paramagnetic phase for all the compounds, the ^{170}Yb absorption Mössbauer spectrum is an axial quadrupolar hyperfine pattern, in agreement with the hexagonal Yb site symmetry. The corresponding hyperfine hamiltonian writes:

$$\mathcal{H}_Q = \alpha_Q[I_z^2 - \frac{I(I+1)}{3}], \tag{5}$$

where $I = 2$ is the spin of the excited nuclear state of ^{170}Yb (the ground nuclear state has spin 0) and α_Q is the quadrupolar hyperfine coupling parameter. The measured quadrupolar hyperfine coupling parameter actually contains two contributions: the $4f$ part, proportional to $\langle Q_{zz}\rangle$, and a contribution from the lattice charges, which is difficult to estimate but whose order of magnitude amounts to 10-20% of the $4f$ contribution, with opposite sign:

$$\alpha_Q = B_Q\langle Q_{zz}\rangle + \alpha_Q^{latt}, \tag{6}$$

with B_Q=0.276 mm/s for ^{170}Yb.

The T=4.2 K quadrupolar spectra in the $YbCu_{5-x}Al_x$ series can all be reasonably fitted using a rather narrow gaussian distribution of α_Q values, with a mean square root deviation amounting to 10% of the central value, which originates from sample inhomogeneities. As can be seen on Fig.**??**, the experimental α_Q values at T=4.2 K decrease as the Al content

x decreases. Using the reasonable assumption that the lattice contribution α_Q^{latt} is constant throughout the $YbCu_{5-x}Al_x$ series, the decrease of α_Q with decreasing x is then due to a decrease of the $4f$ contribution, i.e. of $\langle Q_{zz}(T = 4.2\,K)\rangle$. We will show here that this behaviour can be assigned to the increase of hybridisation as x decreases, and that it enables the values of the Kondo temperature T_K to be determined in the $YbCu_{5-x}Al_x$ series.

According to expression (**??**), the T=0 value of the quadrupolar moment writes (the summation is over the 4 crystal field Kramers doublets, because two Kramers conjugate states have the same quadrupole moment):

$$\langle Q_{zz}(T=0)\rangle = \frac{2\Gamma(1-n_f)}{\pi} \sum_{i=1,4} \frac{\langle Q_{zz}^i\rangle}{T_K + \Delta_i}, \tag{7}$$

where $\langle Q_{zz}^i\rangle$ is the $4f$ quadrupole moment of the ith state. The L_{III}-edge x-ray absorption data [**?**] have shown that the Yb valence, and thus the $4f$ electron count n_f, is constant throughout the series for $x \geq 1.3$, with $n_f \simeq 0.98$. As to the crystal field energies Δ_i, a recent study of the resistivity as a function of pressure in the $YbCu_{5-x}Al_x$ series [**?**] indicates that there is no drastic change of the crystal field splittings as the Al content varies. We can therefore adopt as a reasonable assumption that the CEF interaction is constant throughout the series. Then $\langle Q_{zz}(T=0)\rangle$ is a function of T_K only through Eqns.(**??**) and (**??**), and it is easy to show that the leading term of the derivative $\frac{d\langle Q_{zz}\rangle}{dT_K}$ is proportional to $Q_{zz}^2 - Q_{zz}^1$. Therefore the behaviour of $\langle Q_{zz}\rangle$ as a function of T_K depends on the difference of the $4f$ quadrupole moments of the first excited CEF state and of the ground state. According to Ref. [**?**], the Yb^{3+} ground state ($\Delta_1 = 0$) of the hexagonal crystal field interaction in $YbCu_3Al_2$ is:

$$|\phi_g\rangle = 0.99|\pm\frac{7}{2}\rangle \pm 0.14|\mp\frac{5}{2}\rangle, \tag{8}$$

the first CEF excited state contains the same basis vectors as $|\phi_g\rangle$, but is orthogonal to it, and lies at $\Delta_2 \simeq 100\,K$, and the two other doublets lie at energies higher than 150 K above the ground state. Then: Q_{zz}^1=20.64 and Q_{zz}^2=3.36, and the T=0 hybridised value $\langle Q_{zz}\rangle$ is a decreasing function of T_K ($\frac{d\langle Q_{zz}\rangle}{dT_K} < 0$). Furthermore, as $\Delta_2 \simeq 100\,K$, the T=4.2 K

values of the quadrupolar moment can be considered as the saturated T=0 values. We therefore interpret the experimentally observed decrease of $\alpha_Q(T = 4.2\,K)$ as x decreases in the $YbCu_{5-x}Al_x$ series as due to the increase of the Kondo temperature.

In order to make a more quantitative analysis, we must assess a complete CEF level scheme for the Yb^{3+} ion at its site with hexagonal symmetry. We will model the CEF interaction in the $YbCu_{5-x}Al_x$ series according to the above considerations for the two first excited states, and assume the two remaining states lie at 300 K and 400 K above the ground state. The choice of these two latter energies is rather arbitrary, but their influence on the final result is small. The hexagonal CEF interaction can be expressed in terms of the operator-equivalents O_n^m [**?**]:

$$\mathcal{H}_{hex} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^4 O_6^4. \tag{9}$$

The assumed CEF level scheme for Yb^{3+} corresponds to: $B_2^0 = -10.27$ K, B_4^0=1.3×10^{-2} K, B_6^0=9.7 ×10^{-3} K and B_6^4=1.47×10^{-2} K.

For the less hybridised alloy of the series, $YbCu_3Al_2$, the Kondo temperature was estimated to be around 2.5 K [**?**]. We choose here: T_K= 2.4 K in this compound, which allows to obtain the $\langle Q_{zz}\rangle$ value at T=0 and then the lattice contribution α_Q^{latt} by comparison with the experimental α_Q value according to Eqn(**??**). One obtains: $\alpha_Q^{latt} \simeq -1$ mm/s, which is a correct order of magnitude. Using the assumption that α_Q^{latt} is constant throughout the series, one gets the Kondo temperatures from the measured α_Q values using expression (**??**). The T_K values are represented in Fig.**??** (see also Table I); they increase first moderately as x decreases, then show a rapid upturn for $x \leq 1.5$. Such a variation is expected in the frame of the variational approach, where the Kondo temperature can be shown to grow exponentially as hybridisation increases. For another choice of T_K in $YbCu_3Al_2$ around 2.5 K, the value of α_Q^{latt} would be slightly different, resulting in a small shift of the Kondo temperature values, but with no effect on the main trend of variation of T_K with x.

IV. THE MAGNETICALLY ORDERED PHASE IN THE $YBCU_{5-X}AL_X$ SERIES

The compound $YbCu_3Al_2$ shows antiferromagnetic ordering of the Yb^{3+} moments [**?**,**?**] with T_N=2 K and a saturated magnetic moment of 2.85 μ_B. As x decreases, the increasing hybridisation reinforces the Kondo screening of the Yb^{3+} moment and both the magnetic transition temperature and the saturated moment are expected to decrease [**?**]. We present here Mössbauer and magnetic susceptibility data for the three compounds with x=1.75, 1.7 and 1.6.

A. $YbCu_{3.25}Al_{1.75}$ and $YbCu_{3.3}Al_{1.7}$

The compound with $x = 1.75$ shows a peak of the specific heat at 1 K [**?**] which is attributed to the onset of magnetic ordering. The Mössbauer spectrum at 0.12 K, represented at the top of Fig.**??**, shows the presence of a mean hyperfine field of 2000 kOe at the ^{170}Yb nucleus, hence of a spontaneous Yb^{3+} moment $m \simeq 2\,\mu_B$, using the hyperfine constant for $^{170}Yb^{3+}$ of 1020 kOe per μ_B. This value is reduced with respect to that in $YbCu_3Al_2$ (2.85 μ_B) due to the increased Kondo screening.

In $YbCu_{3.3}Al_{1.7}$, the Mössbauer spectra are quadrupolar hyperfine spectra down to 0.6 K, with however spectral distorsions due to electronic fluctuations in the frequency range 500 MHz – 1 GHz. Below 0.4 K, the spectra change shape and can be interpreted in terms of a static magnetic hyperfine interaction additionally to the quadrupolar interaction. The spectrum at 0.03 K, shown in the middle of Fig.**??**, has a reduced hyperfine splitting with respect to that in $YbCu_{3.25}Al_{1.75}$, corresponding to a hyperfine field of $\sim$700 kOe, i.e. to a spontaneous Yb^{3+} moment of $\sim$0.7 μ_B. This spectrum is however difficult to fit with standard hyperfine lineshapes due to a pronounced inhomogeneity of the linewidths, also visible on the spectrum in $YbCu_{3.25}Al_{1.75}$. This effect will be discussed further in the next section.

In order to determine the magnetic transition temperature more accurately, we performed magnetic susceptibility measurements in a home-made ^{3}He–^{4}He dilution refrigerator with a

SQUID magnetometer, with a field of 5 G, and using both zero-field cooling (ZFC) and field cooling (FC) procedures. The low temperature part of the magnetic susceptibility curve $\chi(T)$ in $YbCu_{3.3}Al_{1.7}$ is shown in Fig.**??**. The transition to the magnetically ordered phase is seen to occur around 0.5 K, in good agreement with the Mössbauer data. The shape of the anomaly in $\chi(T)$ suggests an antiferromagnetic-like ordering. The ZFC and FC curves are distinct below 0.55 K, indicating the presence of irreversibilities in the magnetic behaviour. Below 0.1 K, an upturn is visible on both curves, which could be due to a very small content of paramagnetic "impurity" Yb^{3+} ions not involved in the magnetic order.

B. $YbCu_{3.4}Al_{1.6}$

The Mössbauer spectra in this compound are quadrupolar hyperfine spectra down to the lowest temperature (see the spectrum at 0.04 K at the bottom of Fig.**??**). However, the individual linewidths corresponding to the three nuclear transitions with $\Delta m_I = 0, \pm 1, \pm 2$ reveal a sizeable increase as temperature is lowered below 0.25 K (see Fig.**??**). The lines of a quadrupolar hyperfine spectrum can be broadened by a distribution of electric field gradients, or by the presence of a small hyperfine field which splits or mixes the quadrupolar transitions. As a change in the electric field gradients can be safely ruled out at such a low temperature, we can conclude that the observed broadenings are due to the presence of a small hyperfine field, reaching 100 kOe at 0.04 K. Therefore, the compound $YbCu_{3.4}Al_{1.6}$ shows the onset of a magnetically ordered phase below 0.25 K, with a very small saturated Yb^{3+} moment of $\simeq 0.1\,\mu_B$. The low temperature magnetic susceptibility, measured in a field of 10 G, is shown in Fig.**??**. One observes a deviation from a paramagnetic Curie-Weiss law (dashed line in Fig.**??**) below $\simeq$0.3 K. This anomaly (at 0.3 K) is different from that occuring (at 0.55 K) in $YbCu_{3.3}Al_{1.7}$: it is rather ferromagnetic-like, and furthermore the FC and ZFC curves are practically identical. The temperature at which the susceptibility anomaly occurs is close to that where the Mössbauer linewidths anomaly occurs, and marks the transition to the magnetically ordered state. The alloy $YbCu_{3.6}Al_{1.6}$ lies therefore very close to the

magnetic instability; for a slightly smaller Al content, i.e. for $x = 1.5$, no anomaly in the Mössbauer line broadenings is observed down to 0.06 K. The alloy $YbCu_{3.5}Al_{1.5}$ thus does not show magnetic ordering down to this temperature. For these two alloys very close to the magnetic instability, a clear Non-Fermi liquid behaviour has been evidenced in [**?**] by the specific heat data.

V. THE VARIATIONAL CALCULATION IN THE MAGNETICALLY ORDERED PHASE OF THE $YBCU_{5-X}AL_X$ SERIES

The variational approach is applied here to the calculation of the T=0 spontaneous magnetic moment for $1.6 \leq x \leq 2$. An exchange field $\vec{H}_{ex} = -\lambda\vec{m}$ is introduced and the corresponding exchange interaction is added to the hexagonal CEF interaction of the Yb^{3+} ion. In the presence of the exchange field, the Kondo temperature must be renormalised according to the variational equation:

$$\prod_m (T_K + \Delta_m) = \prod_m [T_K(H_{ex}) + \delta_m(H_{ex})], \tag{10}$$

where T_K and Δ_m are respectively the previously defined zero field Kondo temperature and the CEF energies, and $T_K(H)$ and $\delta_m(H)$ are respectively the renormalised Kondo temperature and the eigenenergies of the total (CEF + exchange) interaction. Using a self-consistent mean field calculation, we computed the T=0 spontaneous moment as a function of the molecular field constant λ, for different values of the Kondo temperature T_K, using expression (**??**) where $O = -g_J\mu_B J_z$ (we assume the spontaneous moment lies along the hexagonal $\vec{c}$ axis, as was demonstrated for x=2 and 1.75 by neutron diffraction measurements [**?**]). The results are shown in Fig.**??**, where the $m = f(\lambda)$ curves were calculated for the T_K values obtained for x=2, 1.75, 1.7 and 1.6. For each value of the Kondo temperature, there is a critical λ value below which no spontaneous moment exists, i.e. below which the Kondo screening of the $4f$ moment overcomes the exchange interaction. Above this critical λ value, the T=0 spontaneous moment steadily grows as λ increases, eventually reaching a value

close to the hybridisation free saturated value $m_0 = g_J \mu_B \langle \psi_g | J_z | \psi_g \rangle$ =3.86 μ_B. By plotting the experimental spontaneous moment values (black squares in Fig.**??**) on the corresponding curves, one obtains the exchange field constant for each value of x (see Table I). The λ value obtained herewith for $x = 2$ (3.7 kOe/μ_B) is in good agreement with that derived in Ref. [**?**] from a fit of the T=1.5 K magnetissation (3.5 kOe/μ_B). The noticeable feature here lies in the fact that the exchange constant values increase as x decreases, i.e. as hybridisation increases. Such a behaviour is expected from the assumption of an RKKY-type exchange between the localised $4f$ moments: then the exchange interaction is expected to vary as the square of the hybridisation parameter Γ/ε_f, where Γ is the already mentioned bare hybridisation width and ε_f is the energy of the $4f$ orbital below the Fermi level. The exchange energy scale can be defined as: $E_{ex} = \lambda m_0^2$, and its values in the $YbCu_{5-x}Al_x$ series are given in Table I. The general trend of variation of the two energy scales T_{ex} and T_K, as well as that of the magnetic transition temperature T_N, with the Al content x agree with Doniach's prediction [**?**] concerning the competition between hybridisation and exchange. As x decreases, T_{ex} and T_K increase, and T_N vanishes for $x \simeq 1.5$ when the two energy scales are of the same magnitude (Fig.**??**).

Using the above described variational calculation, we have also computed the T=0 Mössbauer spectra in the magnetically oredered compounds. This *ab initio* self-consistent calculation was performed with parameters as described in the preceding sections, and assuming that the magnetic moment is parallel to the hexagonal $\vec{c}$ axis. In order to reproduce the inhomogeneous broadenings of the lines, it was necessary to allow for a distribution of distortions from the nominal hexagonal site symmetry. A gaussian distribution of the B_2^0 CEF parameter was introduced; the resulting lineshapes (solid lines in Fig.**??**) agree very well with the experimental spectra with a root mean square deviation $\sigma(B_2^0) = 0.35$ K, amounting to 3.5% of the central B_2^0 value (−10.27 K), for $x = 2$ (not shown, but similar to the fit for x=1.75) and 1.75. For x =1.70, the line broadenings are not exactly reproduced, and for x=1.6, where the magnetic moment is very small, such a procedure does not yield good results. The corresponding solid line in Fig.**??** for $x = 1.6$ is a fit with a standard

hyperfine field lineshape. The inadequacy of the variational calculation for this Al content could be due to the fact that the small Yb^{3+} magnetic moments are not aligned parallel to the $\vec{c}$ axis, but show a more complicated magnetic structure.

VI. CONCLUSION

The variational solution of the impurity Kondo problem has been used to interpret the very low temperature properties of the Yb^{3+} ion in the Kondo lattices $YbCu_{5-x}Al_x$, when the Al content x varies from 1.3 to 2. The single ion Kondo temperatures in this series have been deduced from the T=0 values of the $4f$ quadrupole moment measured by ^{170}Yb Mössbauer spectroscopy. Just above the critical value x_c=1.5 below which the compounds do not show magnetic ordering, we have detected a magnetic transition at T=0.25 K for x=1.6 and at 0.55 K for x=1.70. The values of the magnetic transition temperatures obtained by Mössbauer spectroscopy and by magnetic measurements are in very good agreement. A self-consistent mean field calculation in the frame of the variational approach was used in order to determine the exchange energy from the T=0 spontaneous moment value for $1.6 \leq x \leq 2$. We find that both the Kondo temperature and the exchange energy increase as the $4f$-conduction electron hybridisation becomes stronger, i.e. as x decreases. This behaviour is in good general agreement with the predictions of Doniach's model describing the competition between the Kondo coupling and the exchange interaction for the establishemnt of magnetic ordering in Kondo lattices.

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TABLES

x	1.3	1.4	1.5	1.6	1.7	1.75	2
α_Q (mm/s)	2.75	3.40	3.70	4.00	4.15	4.20	4.35
T_K (K)	31.2	16.4	11.6	7.4	5.1	4.0	2.4
T_N (K)				0.25	0.55	1.0	2.0
m(T=0) $(\mu_B)^a$				0.1	0.7	2.0	2.85
m(T=0) $(\mu_B)^b$						1.2	2.1
λ (kOe/μ_B)				8.3	5.7	5.0	3.7
E_{ex} (K)				8.3	5.7	5.0	3.7

[a] Saturated spontaneous moment values obtained by ^{170}Yb Mössbauer spectroscopy in the present work.

[b] Saturated spontaneous moment values obtained from the neutron diffraction data assuming an antiferromagnetic structure with propagation vector $\vec{k} = (1/2, 1/2, 0)$ and the moments parallel to the $\vec{c}$ axis [**?**].

TABLE I. *Values of the quadrupolar hyperfine coupling parameter α_Q, the Kondo and magnetic transition temperatures, the saturated spontaneous magnetic moment, the exchange constant and the exchange energy in the $YbCu_{5-x}Al_x$ series. The origin of the discrepancy between the spontaneous T=0 moment values derived from ^{170}Yb Mössbauer spectroscopy and neutron diffraction is still an open question.*